Many-Polaron System Confined to a Quantum Dot: Ground-State Energy and Optical Absorption

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Abstract

We find for the first time the ground state energy and the optical absorption spectra for N electrons (holes) interacting with each other and with the longitudinal optical (LO) phonons at an arbitrary electron-phonon coupling strength α in a parabolic confinement potential. A recently developed path integral formalism for identical particles is used in order to account for the fermion statistics. The approach is applicable to closed and open shells. Using an extension of the Jensen-Feynman variational principle, the ground state energy of the N-polarons system in a parabolic confinement potential is analyzed as a function of N and α. A ferromagnetic-to-nonmagnetic transition is shown to occur between states with different total spin of the system in the case of strong electron-phonon interaction. This transition is manifested through the optical absorption spectra and should be experimentally observable. Strong mixing between zero-phonon and one-phonon states is revealed in the optical absorption spectrum, when the confinement frequency parameter is in resonance with the LO phonon frequency ("confinement-phonon resonance"). Moments of the optical absorption spectra are calculated for a N-polaron system in a parabolic quantum dot.

I. INTRODUCTION

The path integral method for indistinguishable particles developed in Refs. 1,2 seems an adequate tool for the investigation of both the equilibrium and the non-equilibrium properties of interacting quantum many-body systems. The advantages of the path integral representation are particularly manifest when investigating systems with a fixed (few or many) number of particles. As demonstrated in Refs. 1,2, the thermodynamical properties of systems with a limited number of particles might deviate substantially from those obtained in the thermodynamical limit.

Electrons confined to a quantum dot can provide a typical example of a system with a fixed number of indistinguishable particles. In recent years, the quantum states and the optical properties of these systems have received considerable attention. Multi-electron states in 3D and 2D quantum dots (without the electron-phonon interaction) have, e. g.,

been treated in Refs. 3–11. It has been shown in Ref. 11 that the interaction between the charge carriers strongly influences the optical spectra of few-particle quantum dots. The bipolaron ground state in quantum dots has been studied in Ref. 12. Cooperative effects for a gas of polarons and bipolarons were treated in Ref. 13 on the basis of a model introduced by Friedberg and Lee[14]. To the best of our knowledge, the effects of the electron-phonon interaction on the optical spectra of multi-electron quantum dots with a fixed number of electrons have hitherto not been investigated theoretically.

Experimentally observed optical absorption spectra of high-T_c cuprates[15–17] are very promising to reveal new manifestations of the interaction between electrons (holes) and the longitudinal optical (LO) phonons. A possible role of polarons in high-T_c superconductivity has been analyzed by several authors (see, e. g., Refs. 18–21). Some aspects of the recently observed optical absorption spectra of high-T_c superconductors[22–24] can be interpreted using the polaron theory[25–27]. It has been shown in Ref. 28 that certain characteristics of the polaron optical absorption at intermediate Fröhlich coupling constant α appear in the aforementioned spectra[15–17]. Bipolaron optical absorption in bulk materials has, e. g., been treated in Refs. 29,30. In view of the relatively high concentration of electrons (holes) in high-T_c superconductors, the development of an all-coupling and all-concentration theory of the optical response of many-polaron systems is an urgent problem.

II. GROUND-STATE ENERGY

In the present communication, we treat the ground state energy and the optical absorption spectra of a multi-electron (multi-polaron) parabolic quantum dot for arbitrary electron-phonon coupling strength, using a recently developed path integral formalism for the quantum statistical treatment of identical particles[1,2] and considering the expected quite large polaron coupling constant in those materials. We consider a system consisting of N electrons in a parabolic confinement potential characterized by the frequency parameter Ω_0. These electrons are assumed to interact with each other and with the LO phonons. The electron subsystem is subdivided into two groups ($N = \sum_\sigma N_\sigma$, where N_σ is the number of electrons with definite spin projection $\sigma = \pm 1/2$).

In the present work the ground state energy of N polarons confined to a quantum dot has been determined for the first time. This calculation has been performed within the *extended Jensen-Feynman variational principle*[1] taking into account the symmetry properties of the electrons with respect to permutations. The validity of this extension of the Jensen-Feynman inequality for systems of indistinguishable particles, which is not obvious, and which is of crucial importance for the present work, has been demonstrated in Ref. 1. We have used an auxiliary model system of particles in a harmonic confinement potential with elastic interparticle interactions as studied in Refs. 1,2. The parameters of the auxiliary model system have been treated as variational parameters. We have worked out the variational procedure to obtain those variational parameters at arbitrary temperature for the physical system of N polarons confined to a parabolic potential. The calculation has been performed for the case of closed shells (i. e. of a non-degenerate ground state) and for the case of open shells.

We have calculated the ground state energy of a parabolically confined many-polaron system as a function of the total spin of the system $S = \frac{1}{2}\left|N_{+1/2} - N_{-1/2}\right|$. For relatively

small polaron coupling constant and all values of $\eta \equiv \varepsilon_\infty/\varepsilon_0$ (ε_∞ and ε_0 are the high-frequency and the static dielectric constants, respectively), except $\eta \ll 1$, the electrons tend to maximally fill up *open* shells with the *same* spin projection (cf. the *first Hund's rule*). In this domain of the parameters α and η, the ground state is obtained when at least one of the numbers $N_{\pm 1/2}$ equals the number of states M_n in a set of shells labelled by the index k from 0 to n (see Ref. 2):

$$M_n = \sum_{k=0}^{n} g_k = \frac{(n+1)(n+2)(n+3)}{6} \quad (n = 0, 1, 2, \ldots), \tag{1}$$

where $g_k \equiv \frac{1}{2}(k+1)(k+2)$ is the degeneracy of the kth energy level of a three-dimensional oscillator. For $n = 0, 1, 2, 3 \ldots$ the values of the number M_n are $1, 4, 10, 20 \ldots$.

This filling scheme is *broken* for strongly polar substances ($\eta \ll 1$), where we find that the lowest variational energy corresponds to the minimal possible total spin ($S = 0$ for N even, and $S = \frac{1}{2}$ for N odd), or for $\alpha \gg 1$ (except the region $\eta \ll 1$), where the lowest variational energy corresponds to the maximal possible total spin ($S = N/2$).

In Table 1, the variational ground state energy per particle E_0/N is shown for $N = 20$ polarons in a quantum dot with confinement frequency parameter $\Omega_0 = 0.5$ at $\alpha = 5$, for different values of η.

Table 1. The variational ground state energy per particle E_0/N (in units of the LO phonon energy $\hbar\omega_{\rm LO}$) for different η for $N = 20$ polarons in a quantum dot with the confinement frequency parameter $\Omega_0 = 0.5$ (in units of $\omega_{\rm LO}$) at $\alpha = 5$.

η	$E_0/N \left(\begin{smallmatrix} N_{+\frac{1}{2}} = 20, \\ N_{-\frac{1}{2}} = 0 \end{smallmatrix} \right)$	$E_0/N \left(\begin{smallmatrix} N_{+\frac{1}{2}} = 10, \\ N_{-\frac{1}{2}} = 10 \end{smallmatrix} \right)$
0.01	-3.9404	-4.1078
0.081	-2.0867	-2.0873
0.082	-2.0626	-2.0620
0.4	5.8487	6.0482

As follows from this table, a ferromagnetic-to-nonmagnetic transition takes place at a value of η in the interval between 0.081 and 0.082. It will be analyzed below how this transition influences the optical absorption spectra.

III. OPTICAL ABSORPTION SPECTRA

In order to investigate the optical properties of the confined many-polaron system, we use the formalism developed in Refs. 25–27. An alternative derivation can be found in Ref. 30. Within this technique, the absorption spectrum for a many-polaron system in a parabolic confinement potential is given by the expression

$$\Gamma(\omega) \sim \frac{-{\rm Im}\chi(\omega)}{\left[\omega - \Omega_0^2/\omega - {\rm Re}\chi(\omega)\right]^2 + \left[{\rm Im}\chi(\omega)\right]^2}, \tag{2}$$

where the memory function is

$$\chi(\omega) = \sum_{\mathbf{q}} \frac{2|V_{\mathbf{q}}|^2 q^2}{3N\hbar\omega} \int_0^\infty \left(e^{i\omega t} - 1\right) \mathrm{Im}\left[T^*_{\omega_{\mathrm{LO}}}(t) \left\langle \rho_{\mathbf{q}}(t)\, \rho_{-\mathbf{q}}(0)\right\rangle_{S_M}\right] dt. \tag{3}$$

The function $T_\omega(t) = \cos\left[\omega\left(t - i\beta/2\right)\right] / \sinh\left(\beta\omega/2\right)$ with $\beta = \hbar/k_B T$ describes the phonon dynamics. The time-dependent correlation function $\left\langle \rho_{\mathbf{q}}(t)\, \rho_{-\mathbf{q}}(0)\right\rangle_{S_M}$ is the path integral average for an auxiliary model action functional S_M of N identical electrons and N_B identical fictitious particles (simulating the influence of the LO phonon bath). As emphasized above, the parameters of the auxiliary model system are determined using the variational procedure for the physical system of N confined polarons under consideration. Note that, formally, the term Ω_0^2/ω plays a role similar to the cyclotron frequency ω_c in the theory of the cyclotron resonance of polarons[31]. In the zero temperature limit, we have obtained the following analytic expression for the memory function:

$$\begin{aligned}
\chi(\omega) = {} & \frac{2\alpha m^*}{3\pi N\omega}\left(\frac{\omega_{\mathrm{LO}}}{A}\right)^{3/2} \sum_{p_1=0}^{\infty}\sum_{p_2=0}^{\infty}\sum_{p_3=0}^{\infty} \frac{(-1)^{p_3}}{p_1!p_2!p_3!}\left(\frac{a_1^2}{N\Omega_1 A}\right)^{p_1}\left(\frac{a_2^2}{N\Omega_2 A}\right)^{p_2}\left(\frac{1}{NwA}\right)^{p_3} \\
& \times \left\{\sum_{m=0}^{\infty}\sum_{n=0}^{\infty}\sum_{\sigma} N_{m,\sigma}\left(1 - N_{n,\sigma}\right)\left[\frac{1}{\omega - \omega_{\mathrm{LO}} - \left[p_1\Omega_1 + p_2\Omega_2 + (p_3 - m + n)\, w\right] + \mathrm{i}\varepsilon}\right.\right. \\
& \left. - \frac{1}{\omega + \omega_{\mathrm{LO}} + p_1\Omega_1 + p_2\Omega_2 + (p_3 - m + n)\, w + \mathrm{i}\varepsilon} + 2\frac{\mathcal{P}}{\omega_{\mathrm{LO}} + p_1\Omega_1 + p_2\Omega_2 + (p_3 - m + n)\, w}\right] \\
& \times \sum_{l=0}^{m}\sum_{k=n-m+l}^{n} \frac{(-1)^{n-m+l+k}\,\Gamma\left(p_1 + p_2 + p_3 + k + l + \frac{3}{2}\right)}{(k)!\,(l)!} \\
& \times \left(\frac{1}{wA}\right)^{l+k}\binom{n+2}{n-k}\binom{2k}{k-l-n+m} + \left[\frac{1}{\omega - \omega_{\mathrm{LO}} - (p_1\Omega_1 + p_2\Omega_2 + p_3 w) + \mathrm{i}\varepsilon}\right. \\
& \left.\left. - \frac{1}{\omega + \omega_{\mathrm{LO}} + p_1\Omega_1 + p_2\Omega_2 + p_3 w + \mathrm{i}\varepsilon} + 2\frac{\mathcal{P}}{\omega_{\mathrm{LO}} + p_1\Omega_1 + p_2\Omega_2 + p_3 w}\right]\sum_{m=0}^{\infty}\sum_{n=0}^{\infty}\sum_{\sigma,\sigma'} N_{m,\sigma}N_{n,\sigma'}\right. \\
& \left. \times \sum_{k=0}^{n}\sum_{l=0}^{m} \frac{(-1)^{k+l}\,\Gamma\left(p_1 + p_2 + p_3 + k + l + \frac{3}{2}\right)}{k!l!}\left(\frac{1}{wA}\right)^{k+l}\binom{n+2}{n-k}\binom{m+2}{m-l}\right\},
\end{aligned} \tag{4}$$

where $\varepsilon \to +0$, $\mathcal{P}$ denotes the principal value, $A \equiv \left[\sum_{i=1}^{2} a_i^2/\Omega_i + (N-1)/w\right]/N$. Ω_1, Ω_2 and w are the eigenfrequencies of the model system (Ω_1 is the frequency of the relative motion of the center of mass of the electrons with respect to the center of mass of the fictitious particles; Ω_2 is the frequency related to the center of mass of the entire model system; w is the frequency of the internal degrees of freedom), a_1 and a_2 are the coefficients of a canonical transformation which diagonalizes the model Lagrangian, and $N_{n,\sigma}$ is the number of electrons with spin projection σ in the nth single-particle level (shell).

IV. DISCUSSION OF RESULTS

In Fig. 1, the optical absorption spectra of a many-polaron system in a quantum dot with parabolic confinement potential are plotted for $\Omega_0 = 0.5$ (all frequencies are measured in units of ω_{LO}), $\alpha = 1$ and for $N = 5, 10, 14$ (panels a, b, c, respectively). Due to the confinement in all three dimensions, the electron motion is fully quantized. Hence, when

a photon is absorbed, the electron recoil can be transferred only by discrete quanta. As a result, the absorption spectrum consists of a series of δ-like peaks as distinct from the absorption spectrum of a bulk polaron. In this and subsequent figures, the height of each peak represents its intensity.

Fig. 2 shows, for reference, the optical absorption spectrum for a *single* polaron confined to a quantum dot with $\Omega_0 = 0.2$ for $\alpha = 5$, and reveals several essential elements. Following the nomenclature of Refs. 25,26 we distinguish a central (zero-phonon) peak, peaks due to transitions to the relaxed excited state (RES), and peaks associated with transitions to the Franck-Condon state (FC). Also it is worth mentioning that there is the "one-phonon shoulder" in the optical absorption spectrum with threshold at $\omega \sim 1$. The envelope of the absorption spectrum for a polaron in a quantum dot is very similar to the bulk polaron absorption spectra for the same α, as first obtained in Ref. 25. It follows that the obtained optical absorption spectra for a polaron confined to a quantum dot are consistent with those for a bulk polaron[25,26].

As follows from Figs. 1 and 2, the δ-like "central peak" for a system of polarons, confined to a quantum dot with parabolic confinement potential, is positioned at $\omega \sim \Omega_0$. Without the electron-phonon interaction, the "central peak" for a system of electrons, confined to a quantum dot with parabolic confinement, is exactly at the confinement parameter: $\omega = \Omega_0$.

Fig. 3 shows the evolution of the absorption spectrum as a function of the confinement parameter Ω_0, for $\Omega_0 = 0.5$, $\Omega_0 = 0.8$, $\Omega_0 = 1$ and $\Omega_0 = 1.2$. This calculation is performed for $N = 4$, $\alpha = 3$, $\eta = 0.3$. Near $\Omega_0 = 1$, the zero-phonon and the one-phonon peaks are of comparable oscillator strength. A formal analogy (particularly manifest for sufficiently small α) exists between the resonance condition $\Omega_0 = 1$ discussed here and the wee-established magnetophonon resonance around $\omega_c = 1$ (Refs. 32,33). We suggest the designation "confinement-phonon resonance".

Fig. 4 illustrates the ferromagnetic-to-nonmagnetic transition induced by the increase of the electron-phonon interaction for a many-polaron system confined to a quantum dot. In this figure, optical absorption spectra are plotted for $N = 20$ polarons in a quantum dot with confinement parameter $\Omega_0 = 0.5$, and with electron-phonon coupling constant $\alpha = 5$. The parameter η varies from $\eta = 0.01$ to $\eta = 0.4$, and we find that, as a consequence, the total spin of the ground state jumps from $S = 0$ to $S = 10$. As seen from Table 1, for $\eta \leq 0.081$, the ground state with $S = 0$ is energetically favorable. The optical absorption spectra for this case are shown on panels *a* ($\eta = 0.01$) and *b* ($\eta = 0.081$) of Fig. 4. A series of pronounced peaks clearly related to the internal polaron excitations (cf. RES of a single polaron in Fig. 2) is seen in the spectrum for the case of a strongly polar substance ($\eta = 0.01$). With increasing η, the relative intensity of these "RES" (many-polaron) peaks with respect to that of the zero-phonon peak decreases as is seen from Fig. 4. This effect is due to weakening of the electron-phonon interaction with increasing parameter η.

In the specific case under consideration ($N = 20$), when η varies from $\eta = 0.081$ to $\eta = 0.082$, the total spin abruptly changes from $S = 0$ [$N_{1/2} = 10$, $N_{-1/2} = 10$] to its maximal value $S = 10$ [$N_{1/2} = 20$ (0), $N_{-1/2} = 0$ (20)], i. e., the electrons become fully spin-polarized. Such a jump in the magnitude of the total spin can also be realized by varying the confinement parameter at fixed α and η. Comparing the optical absorption spectra in panels *b* and *c* of Fig. 4, one observes that the optical absorption spectrum as a whole shows an abrupt change at the ferromagnetic-to-nonmagnetic transition, though the shift of the

most intense phonon sidebands towards lower frequencies (with increasing η) continuously proceeds when η passes through the value 0.081. Note also the shift to lower frequencies of the most intense phonon sidebands (again related to the "many-polaron RES") in Fig. 4*b* compared to Fig. 4*a*.

V. MOMENTS OF THE OPTICAL ABSORPTION SPECTRA

We have calculated (both for closed-shell and open-shell systems) the first frequency moment M_1 of the optical absorption spectrum for a N-polaron system confined to a quantum dot with parabolic confinement potential

$$\langle\omega\rangle \equiv \frac{\mu_1}{\mu_0} = \frac{\int_0^\infty \omega\Gamma\left(\omega\right) d\omega}{\int_0^\infty \Gamma\left(\omega\right) d\omega} \tag{5}$$

with $\Gamma\left(\omega\right)$ the optical absorption coefficient, and the parameter $\sigma \equiv \sqrt{\left\langle \left(\omega - \langle\omega\rangle\right)^2 \right\rangle}$, where

$$\left\langle \left(\omega - \langle\omega\rangle\right)^2 \right\rangle = \left\langle \omega^2 \right\rangle - \langle\omega\rangle^2 \tag{6}$$

is the normalized second frequency moment of the optical absorption spectrum.

The results for the first frequency moment of the optical ansorption spectrua for N polarons in a quantum dot with parabolic confinement are shown in Fig. 5*a*. The first frequency moment is plotted as a function of the effective density $N/\mathcal{V}$, where $\mathcal{V} = (4\pi/3)\left(\hbar/2m\Omega_0\right)^{3/2}$. The points corresponding to definite N are shown by filled circles. The number of particles ranges from $N = 1$ to $N = 20$. The points indicated by arrows are related to the closed-shell systems.

The function $\langle\omega\rangle\left(N/\mathcal{V}\right)$ turns out to be non-monotonous. There is a maximum of the first frequency moment $\langle\omega\rangle$ at $N = 2$. The first frequency moment has a minimum when the number of particles takes the value $N = 14$ corresponding to the closed-shell system ($N_{+1/2} = 10$, $N_{-1/2} = 4$).

By its general trend, the first frequency moment of the optical absorption spectrum as a function of the effective density strikingly resembles the first moment of optical conductivity spectrum in $Nd_{2-x}Ce_xCuO_{4-y}$ recently observed experimentally (Fig. 5*b* and Ref. 34). Note that our theory describes a 3D confined system, while the experiment relates to a quasi-2D translationally invariant system. Therefore, the theory can be expected to reveal only a *qualitative trend* of the first frequency moment as a function of concentration in comparison with experiment.

VI. CONCLUSIONS

We have presented the ground state energy and the optical absorption spectra calculated for a system of N polarons in a parabolic confinement potential for any strength of the polaron coupling. Path integral formalism for the quantum statistical physics of indistinguishable particles[1,2] has allowed to develop the variational procedure[1] for the ground state energy for a finite number of polarons. For the first time, the ground state energy and

the optical absorption spectra have been analyzed for N electrons (holes) interacting with each other and with the longitudinal optical (LO) phonons at an arbitrary electron-phonon coupling strength α to a parabolic confinement potential. A new type of transition for N polarons confined in a parabolic potential (ferromagnetic-to-nonmagnetic transition) is found between states with different total spin, which is related to the competition between the *Coulomb repulsion* and the *phonon-mediated attraction* between the electrons. For relatively weak polaron coupling constant, the electrons are shown to maximally fill up shells with the same spin projection (cf. the first Hund's rule). This filling scheme is demonstrated to be broken for strongly polar substances ($\eta \ll 1$), where we find that the lowest variational energy corresponds to the minimal possible total spin. The present analysis has been executed for closed-shell and open-shell systems.

The optical absorption spectra have been calculated here using the memory function approach as applied to path integrals for a many-polaron system confined to a quantum dot with different number of polarons. The dependence of the optical absorption spectra on the confinement parameter Ω_0 reveals a resonant behavior for $\Omega_0 \approx 1$, especially if α is small. The polaron RES[25,26] are seen to also influence the optical absorption spectra of N confined polarons.

We have analysed also the first frequency moment of the optical absorption spectrum for a N-polaron system in a parabolic quantum dot for both closed-shell and open-shell systems.

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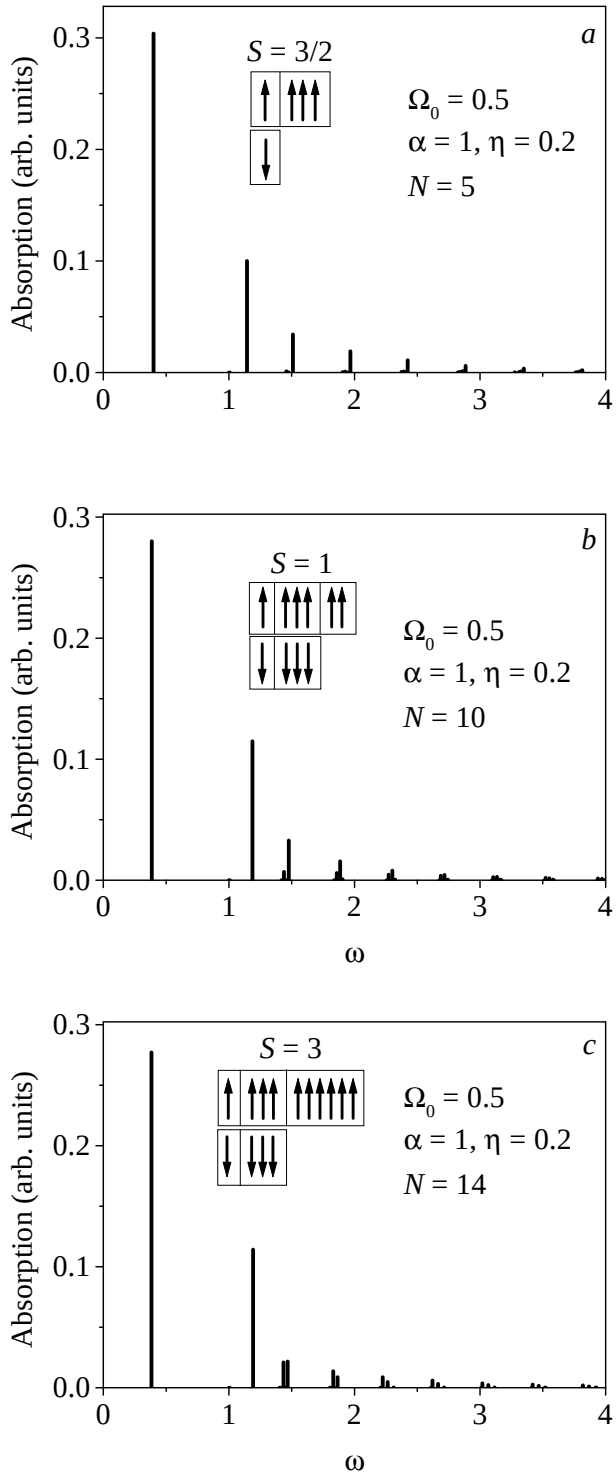

Fig. 1. Optical absorption spectra of N polarons (N = 5,10,14) in a quantum dot with parabolic confinement potential for $\alpha = 1$, $\eta = 0.2$. The confinement parameter is $\Omega_0 = 0.5$ (in units of ω_{LO}). In all figures, the boxes with the arrows inside show the filling of the shells. The up and down arrows represent spin projections.

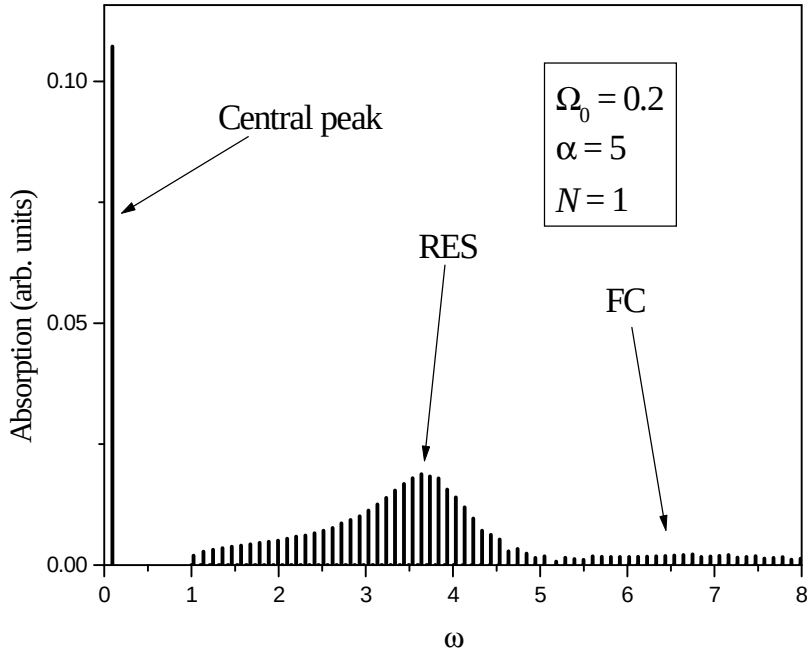

Fig. 2. Optical absorption spectrum of one polaron in a quantum dot with parabolic confinement potential for the confinement parameter $\Omega_0 = 0.2$ and for $\alpha = 5$.

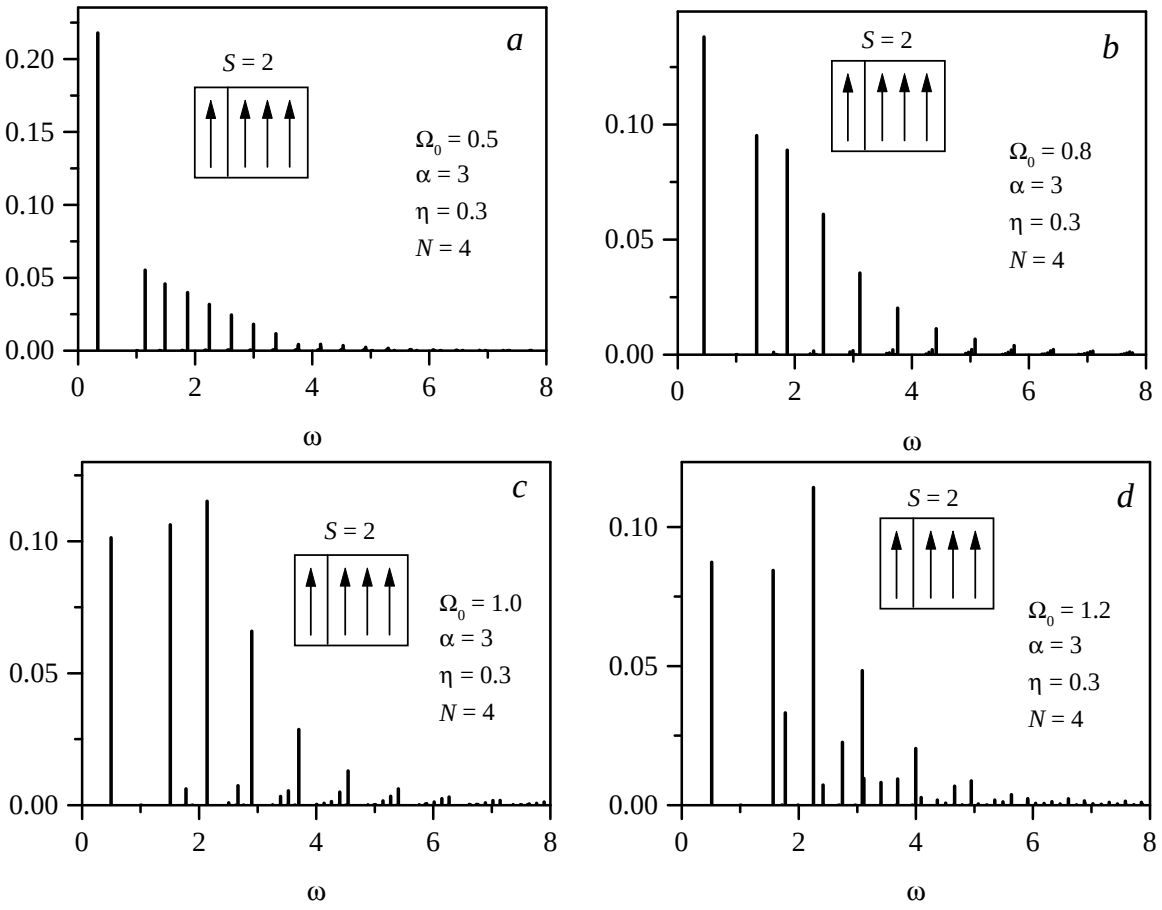

Fig. 3. Evolution of the optical absorption spectrum of the *N*-polaron system in a quantum dot with parabolic confinement potential as the confinement parameter changes from $\Omega_0 = 0.5$ to $\Omega_0 = 1.2$. The following parameters are chosen: $N = 4$, $\alpha = 3$, $\eta = 0.3$. The total spin of the ground state is $S = 2$.

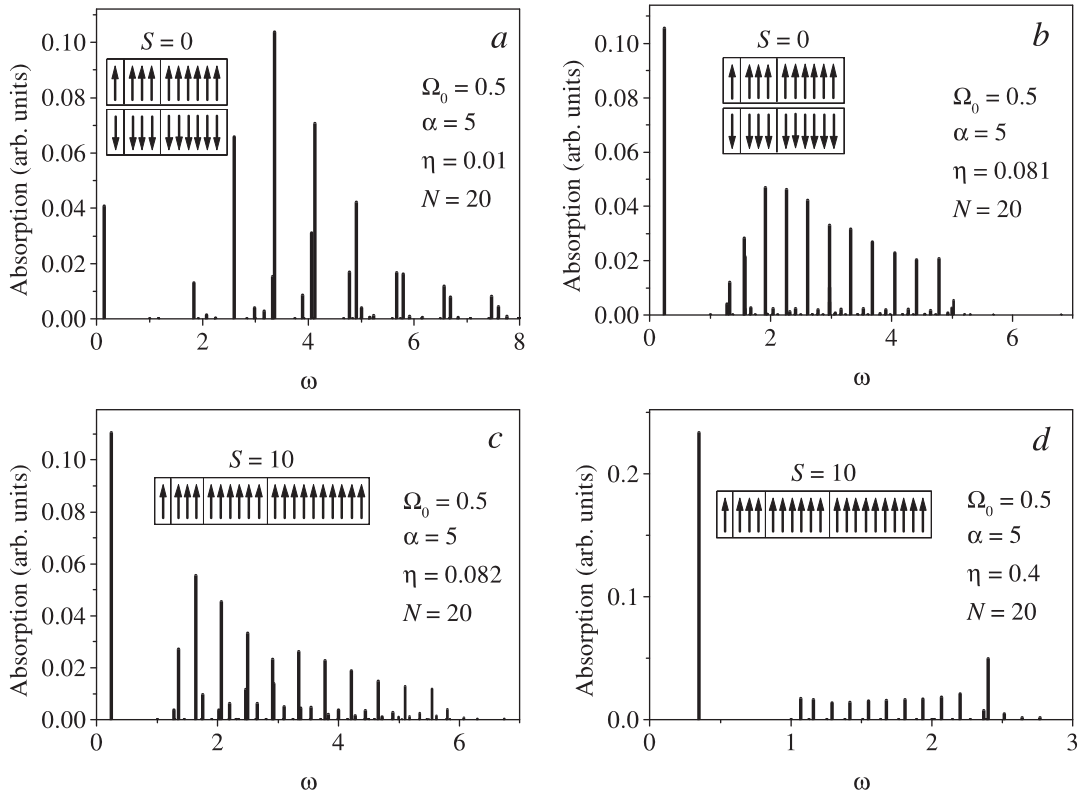

Fig. 4. Optical absorption spectra of an interacting many-polaron system in a quantum dot with parabolic confinement potential for $N = 20$, $\alpha = 5$, and for different values of the parameter η. The confinement parameter is $\Omega_0 = 0.5$.

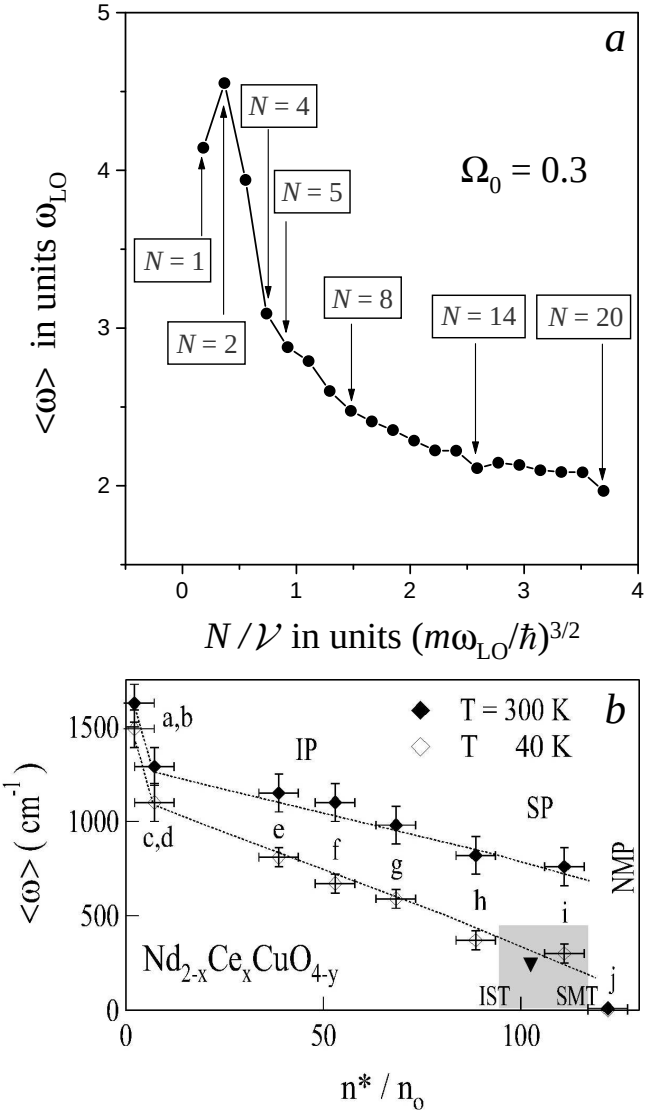

Fig. 5. *Panel a*: First frequency moment <ω> of the optical absorption spectra for N polarons (N = 1 to 20) in a 3-dimensional QD with parabolic confinement for $\alpha = 6$, $\eta = 0.05$, $\Omega_0 = 0.3$. The points indicated by arrows correspond to the closed-shell systems. The parameter $\mathcal{V} = (4\pi/3)(2m\Omega_0\omega_{LO}/\hbar)^{3/2}$. For ω_{LO} = 506 cm^{-1} [35], and $m = m_0$ [34], $(m\omega_{LO}/\hbar)^{3/2} = 7.48\times10^{20}$ cm^{-3}.

Panel b: First frequency moment of the measured polaron optical conductivity spectra at 300 K (open squares) and 40 K (full circles) for all samples except sample "i" (measured at 10 K) vs the normalized effective carrier concentration n^*/n_0 (where n_0 is the carrier concentration in the less doped sample "a"). The superconducting phase (SP) is shown by a shaded area. IST roughly indicates the transition from the insulating phase (IP) to the superconducting one. SMT approximately indicates the transition from the superconducting phase to the normal metal phase (NMP). The LO phonon frequencies ω_{LO} for $x = 0.15$, $y = 0$ at 295 K are 596, 506, 490 cm^{-1} [35]. According to the value of the plasma frequency from [35], the concentration corresponding to $n^* = n_0$ is estimated to be ~ 10^{19} cm^{-3}. By courtesy of P. Calvani.

As discussed in the text, comparison between theory and experiment reveals only qualitative trends of <ω> as a function of concentration.